Electrical Conductivity of Tungsten in a Continuous Transition from Condensed to Gaseous State[1]

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A pulse heating technique is developed that makes it possible to investigate the transition of a metal from condensed to gaseous state while maintaining almost uniform temperature and pressure distributions in a sample. By means of the technique the electrical conductivity of tungsten was measured in a process during which a pressure in the range of 30 – 90 kbar was applied to the sample and its density decreased from the standard solid density down to a density of 15 to 20 times lower. Since the pressures are substantially higher than the critical pressure the transition from condensed to gaseous state was continues. The earlier obtained results have shown that along isobars in the range of 30 - 60 kbar the density dependence of conductivity changes radically at a certain density value (at which it has a pronounced knee). At the knee, the density is approximately ten times lower than standard solid density, and the internal energy is about two times the sublimation energy. The dependence of the conductivity near the knee becomes smoother when the pressure increases. In the paper we present new results on the conductivity of tungsten at the pressures up to 90 kbar. It is shown that the knee becomes remarkably flatter and smoother at these pressures compared with the corresponding low pressure results. Nevertheless the main features of the conductivity dependence observed at the low pressures persist at the maximal pressure applied.

KEY WORDS: metal-non-metal transition; electrical conductivity; tungsten; strongly-coupled plasma; exploding wires; liquid metal.

1. INTRODUCTION

The problem of transitions of metals from condensed to gaseous state was raised in the well-known paper by Zel'dovich and Landau [1]. They noticed that there is no qualitative difference between metallic and dielectric states at finite temperatures and therefore, transition from one state to another may be continuous. The question then arises as to the character of the phase diagrams of metals in the region where the metal-to-dielectric transition takes place. The authors predicted that in general metals should undergo two separate first-order phase transitions, specifically, liquid-to-vapor and metal-to-dielectric transitions with two different critical points. This question in the case of refractory metals remains unanswered because of the difficulties in performing precise measurements at high pressures and temperatures.

In a number of papers, attempts were made to measure the electric conductivity of metals in a wide density range. Some modifications of the exploding wire technique were used in [2, 3, 4] to measure the conductivity of the strongly coupled metallic plasmas. The electric conductivity was determined assuming that the temperature, pressure, and other quantities are uniformly distributed over the column formed by an exploding wire (or a foil [4]). At the initial stage of the heating process during which the wire evaporates the sample was substantially nonuniform. It is assumed by the authors that the nonuniformities disappear to the later stage of the process. No direct evidences of such behavior are provided in the mentioned works.

This was the main motivation for us to study the exploding wire dynamics when

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a sample (a wire or a foil), placed in a condensed dielectric medium (water, glass capillary, plastic coatings) is heated by an intense electrical current pulse. In this study a one-dimensional magneto-hydrodynamic model was developed describing the hydrodynamic processes, which take place within a metal undergoing melting, evaporation, and ionization. The computational study has revealed the influence of different factors on the spatial distributions of physical quantities in the sample subjected to the pulse heating. Basing on this study, an original technique was proposed in [5] realizing the transition of a metallic sample from condensed to gaseous state while maintaining the uniform distributions of the temperature and pressure in it.

In the paper [6], we reported the results of experiments utilizing this technique to measure electric conductivity of tungsten in a process during which the pressure in the sample was maintained at a level of 30 – 60 kbar and the density decreased from the normal solid density down to a density of 15 to 20 times lower. Since such pressures are several times higher than the critical pressure for the liquid-to-vapor phase transition (the critical pressure of tungsten is within 11 – 13 kbar [7]), evaporation did not manifest itself in these experiments. Therefore the transition from condensed to gaseous state was continues in these experiments. The point is that the volume evaporation (boiling) inevitably leads to the formation of nonuniform temperature and density distributions in the sample [7].

The experimental results presented in [6] show that, along the isobars $P = 30$ - 35 kbar, the density dependence of the conductivity of tungsten changes radically at a certain density value, at which it has a pronounced knee[1]. At the knee, the density is approximately ten times lower than the standard solid state density, and the internal energy is about two times the sublimation energy. For the $P = 60 – 65$ kbar isobars, the density dependence of conductivity near the knee becomes smoother. In the paper we present results on the conductivity of tungsten obtained at the pressures up to 90 kbar. It is shown that at the maximal pressure applied the knee becomes flatter. The results obtained are compared with the literature data.
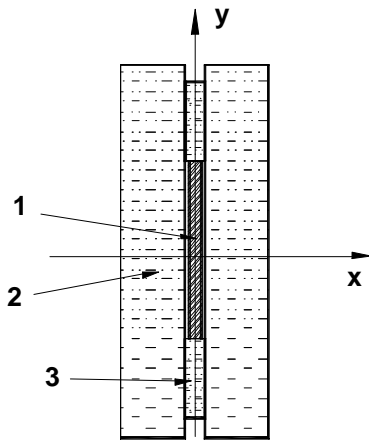
Fig. 1. Transverse cross-section of experimental assembly: foil strip (1); glass plate (2); mica plate (3). The current flows in a direction perpendicular to the plane of the figure.

3. DESCRIPTION OF THE PULSE HEATING TECHNIQUE

The experiments were carried out with tungsten foil strips with a thickness of 20–22 μm, width of 1.5–3.0 mm and length of about 1.0 cm. A foil sample was placed between two polished sapphire (or glass) plates having a thickness of 5–7 mm, width of 10 mm, and length of 10.0–11.5 mm. The side slits were filled in by two mica plates (See, Fig. 1). The experimental assembly was made so that the gaps between the sample and the plates were minimal. Due the roughness of the surfaces the gap was usually within 5–7 μm. The pulse heating of the sample was accomplished by discharging a 72-μF capacitor bank at a charging voltage of about 18 kV.

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[1] Prof. A. W. DeSilva has proposed to use the word "knee" to characterize the effect.

The temporal dependencies of the current through the sample and the voltage drop across its length were measured. The resistance of the sample and the Joule heat dissipated in it were determined from these two measured quantities. The electric conductivity was calculated according to the formula:

$$\sigma = \frac{l}{SR} \qquad (1)$$

where l is the sample length, R is its resistance, and S is its cross-sectional area, i.e. the product of the sample width h and thickness $d(t)$. The thickness as a function of time was calculated using a hydrodynamic model and assuming the length and width are constant.

Basing on the numerical modeling results in the framework of the one-dimensional magneto-hydrodynamic model [6, 7] the basic parameters of the experiments presented here were chosen so that the sample remains homogeneous during the pulse heating process. A detailed explanation of the work is discussed in [6]. The calculations of the sample thickness for the homogeneous heating experiments can be performed for the prescribed temporal dependencies of the Joule heat released and the electrical current pulse taken from the corresponding experiments. In this case, i.e. for the homogeneous heating the hydrodynamic equations of the model [6, 7] should be solved only (the laws of conservation of the mass, linear momentum and energy) the right hand parts of which are some given functions of time (see, [6]).

Thus, the conductivity is obtained using an equation of state model only. The quality of the equation of state utilized in the paper was carefully analyzed in [7] for the region of the liquid-to-vapor phase transition (up to the critical point). The ionization effects were described in the average atom approximation [8]. The corresponding terms were added to the thermodynamic functions used in [7].
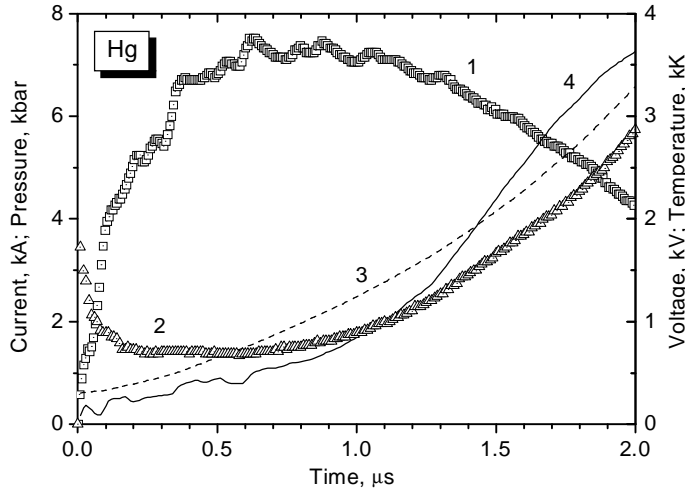
Several experiments were performed to prove that the sample remains homogeneous. In particular, the experiments with the different sample widths were carried out. Every experiment was repeated at least two times to get the information about the reproducibility and uncertainty for the measured quantities. These experiments have shown that the current and voltage are measured with an uncertainty less than 5%. The resistance as a function of the Joule heat released for different experiments with all parameters kept constant scatters less than 10%. To prove that the complete procedure used for determination of conductivity (when some quantities are measured and other are calculated) is self-consistent a special investigation was carried out. Several experiments were performed in which a thermodynamic state in the P-T plane was achieved by substantially different ways. As it was shown in [6] the values of the resistivity at this state obtained were within the experimental uncertainty of 10%.

Nevertheless, a systematic error may exist in such a procedure when the current and voltage drop are only measured and the other quantities including the sample thickness and pressure are calculated. To answer the question some experiments with mercury were performed.

4. COMPARISON WITH THE STEADY-STATE DATA
The results on the mercury conductivity were obtained by means of the pulse heating technique presented in this paper. The main reason to investigate mercury was to compare our results with the literature steady-state data in the metal-to-dielectric transition domain. In the case of mercury this domain is located at low enough temperatures and pressures and therefore, there are reliable literature data on the equation of

state information and the conductivity as well. The thermodynamic information along with the critical point parameters were used to fit the adjustable parameters of the equation of state model. The fitting procedure is described in [7]. As a matter of fact, after this fitting the equation of state was able to reproduce the reliable literature data on the critical point parameters, the thermal expansion coefficient and the specific heat at the constant pressure with a 3-5% discrepancy.

Fig. 2. Temporal dependences of the basic measured (marks) and calculated quantities (lines) for an experiment with Hg sample: current (1); resistive part of the voltage drop (2); temperature (3); pressure (4).
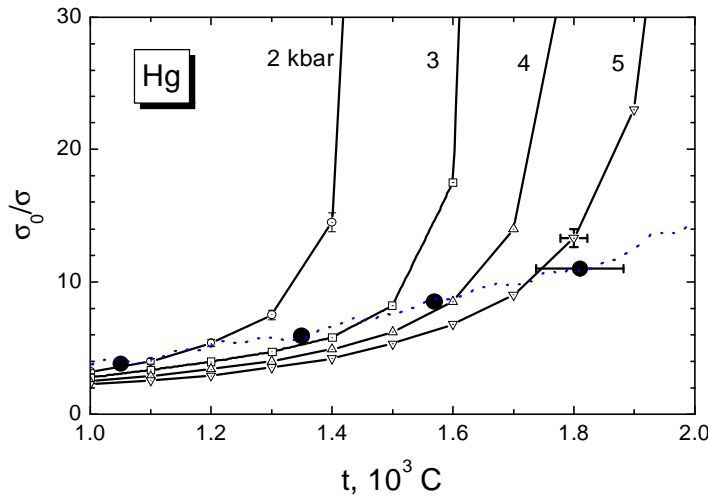
Fig. 3. Relative resistivity versus temperature. Open marks connected by solid lines, isobars of the steady state experiments [9]; blue dotted line, this work; closed circles designate the points at which the pressure values of 2, 3, 4, and 5 kbar were achieved in this work pulse heating experiment.

In the experiments on mercury the samples with a thickness of 60 μm, width of about 3 mm, and length of 10 mm were used. A cavity formed by two polished glass plates having a thickness of 5 mm, width and length of 10 mm and two thin mica plates was filled in by liquid mercury. The pulse heating of the sample was accomplished by discharging the capacitor bank at a charging voltage of about 10 kV.

The temporal dependences of the measured and some calculated quantities are presented in Fig. 2. The ratio of the resistivity (σ^{-1}) to its value at the standard conditions versus temperature is presented in Fig. 3. As one can see our measurements are in a good agreement with the steady-state data [9]. This conclusion gives a confidence that the technique used is able to obtain the reliable data on the conductivity in the metal-to-dielectric transition domain.

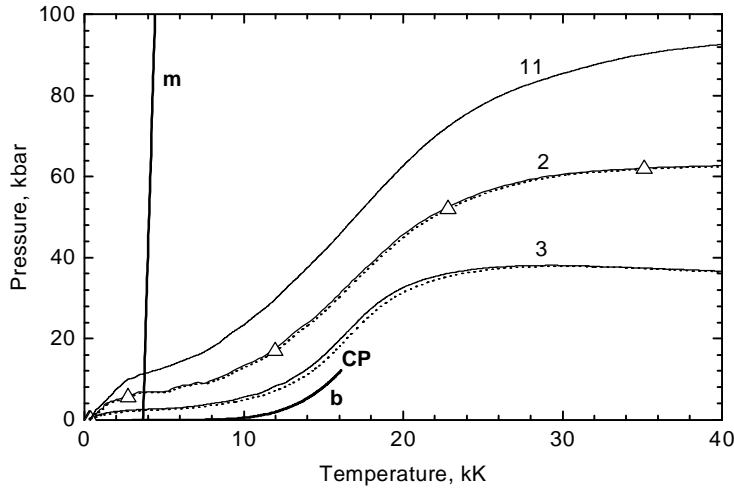### 3. ELECTRICAL CONDUCTIVITY OF EXPANDED TUNGSTEN

The paths of several heating processes realized in our experiments on tungsten in the P-T plane are presented in Fig. 4. The paths were calculated using the mentioned

Fig. 4. Evolution of thermodynamic state of a tungsten foil strip placed between glass plates (experiment 3) and sapphire plates (experiments 2, 11) during the pulse Joule heating process: (m) melting line; (b) boiling curve; (CP) critical point. The triangles refer to times separated by an interval of 200 ns, starting from the time at which the current is switched on.

across the foil sample did not exceed 10% during the entire heating processes. Such nonuniformities are developed mainly due to the pinch-effect and the effect of inertia in the foil sample when the heating power changes.
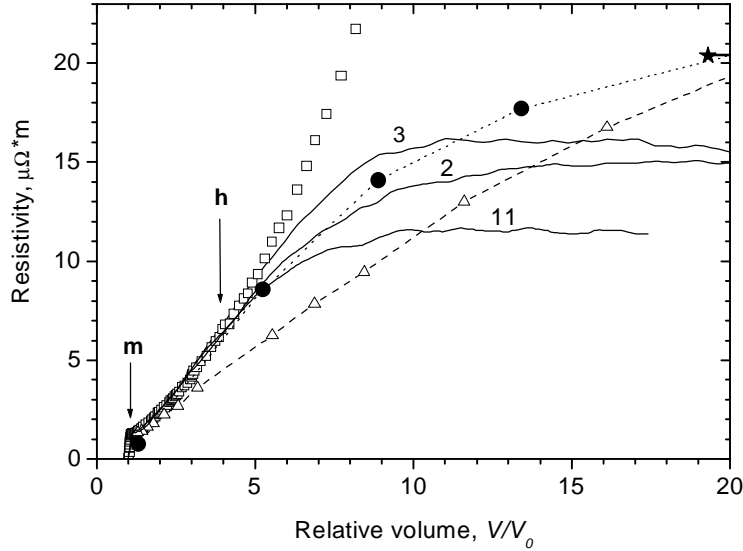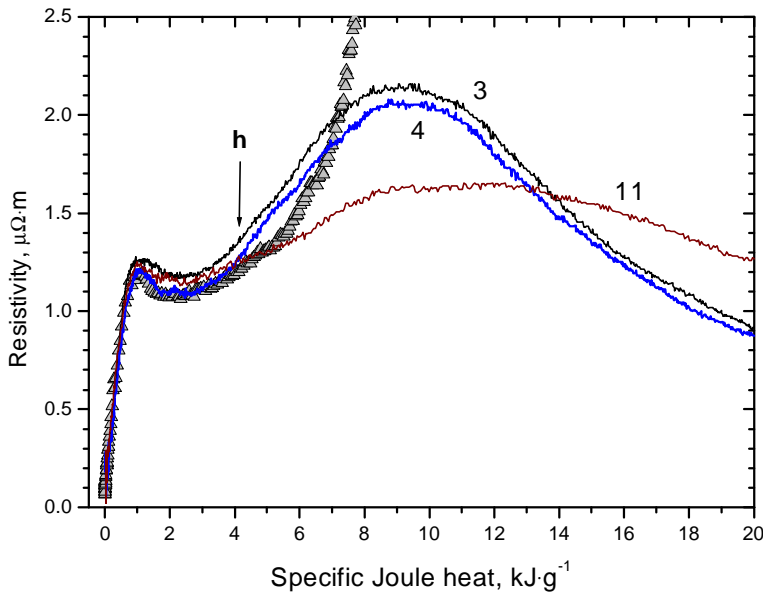

Fig. 5. Resistivity of tungsten versus relative volume: labeled solid lines, this work data; squares, experiment [7]; triangles, experiment [10]; star in the upper right corner, calculation for W (T = 30 kK) [12]. The arrow (m) indicates the melting region; (h) indicates the end of the homogeneous heating stage for experiment [7]. For comparison the first-principle calculations results for Al (T = 30 kK) [11] are also shown (closed circles).

hydrodynamic model for the prescribed temporal dependences of the Joule heat release and current pulse. Each of the three experiments (2, 3 and 11) is illustrated by two trajectories representing two thin layers in the sample: a surface layer and a layer near the symmetry plane. These two layers correspond to two cells from 50-100 cells of the spatial mesh used in these computations. Every layer contains a fixed mass. As follows from Fig. 4, the variations of the temperature and pressure

The corresponding dependences of the resistivity versus relative volume (the volume normalized to the specific volume of solid tungsten under standard conditions) are presented in Fig. 5. As it can be seen, these dependences are different for solid, liquid, and gaseous state. The clearly expressed break designated by the arrow (m) indicates the melting region. In the liquid phase, the resistivity is approximately a linear function of the

specific volume. Such form of the dependence is held up to a relative volume of about 8. Over the range of relative volumes from 8 to 11, the dependence changes its character (experiment 3). For larger relative volumes, the resistivity approaches a nearly constant value. Therefore we can assume that the resistivity dependence changes its form from a metal-like to a plasma-like one at a density which is 8 to 11 times lower then the standard solid density. It should be noted that for relative volumes larger than 5, the heating processes in experiment 3 was nearly isobaric. Thus, we can emphasize that the resistivity changes its density dependence form along an isobar in which it has a pronounced knee in the relative volume range from 9 to 11 (experiment 3). As the pressure increases the dependence becomes smoother and flatter.

Figure 6 shows the resistance of the sample as a function of the specific Joule heat released for experiments 3, 4 and 11. We present these dependences because both of these quantities are measured directly. For convenience in comparing our results with the data obtained in other works, we normalize the resistance by multiplying by the ratio of the initial cross-sectional area of the foil strip to its length: $R^* = R \cdot S_0 / l$. For a homogeneous sample, the resistance so normalized is proportional to the density to conductivity ratio. It can be seen from Fig. 6, that our results agree well with the data obtained in [13, 14], and [7] within the homogeneous heating stage. In [7] the electric conductivity was measured directly at pressures between 10 and 20 kbar at the densities ranging from the standard solid density down to a density of four times lower. The arrow (h) indicates the end of the homogeneous heating stage for those experiments. The distributions of basic quantities across an expanding sample were investigated in [7] by means of hydrodynamic modeling. In Fig. 6 the curve referring to experiment 3 has pronounced maxima in the range of q values from 8 to 10 kJ·g^{-1}. The reproducibility of the data in the region of the maxima is fairly high. To make this evident, we show the data obtained in experiments 3 and 4, which were performed under similar conditions. The two corresponding curves are seen to differ by no more than about 10%. This difference characterizes the accuracy with which the parameters of our experiments are reproduced. It follows from the figure that our results differ from the data obtained in [7] by the same amount.

Fig. 6. Resistivity of tungsten without correction for expansion versus specific Joule heat released: this work data, lines; closed triangles, [7]; open circles, [13]; open squares, [14]. The arrow indicates the end of the homogeneous heating stage for experiment [7].

5. DISCUSSION

At the supercritical temperatures and pressures the density dependence of the electrical conductivity of tungsten changes sharply in a narrow density range in which it has a pronounced knee. When the pressure increases from about 30 to 90 kbar the dependence becomes substantially flatter and smoother. Note that the change of the character of the dependence of the resistivity on the relative volume is directly related to the maximum in the dependence of the resistance on the Joule heat dissipated (See Fig. 6) and, therefore, cannot depend on the accuracy with which the thermodynamic functions of tungsten are described in the corresponding hydrodynamic calculations.

Another interesting feature in the resistivity behavior as a function of density is the following. For a given pressure a characteristic density value exists above which the resistivity merges with a limiting envelope of the resistivity isobars. It can be clearly seen in Fig. 5 that the relative volume at which this takes place depends on pressure. In the work [11] the same behavior is discussed for conductivity of aluminum along isothermals. Examining the band structure of the metal when its density is lowered the authors [11] have revealed formation of a gap in the electronic density of states near the Fermi energy at a certain density value. The data obtained for aluminum in [11] show a similar behavior compared with this work data obtained for tungsten. The value of the relative volume at which the curve representing the data [11] changes its slope correlates well with the position of the knee observed in this work. The reason to compare our data with the aluminum data is based on the similarity in the dependence of the conductivity versus atomic number density observed for several metals including tungsten and aluminum in [3]. In [11] it is noticed that in the liquid phase at the densities larger than the characteristic density at which the gap is formed the conductivity very weakly depends on the temperature but strongly on density: $\sigma \propto \rho^{7/3}$. This work results show that the tungsten conductivity is roughly linearly dependent on density in a wide density range (from the melting point density down to a density of 6-7 times less). More careful analysis shows that the conductivity dependence can be approximated by $\sigma \propto \rho^n$ only for a density range from the melting point density down to a density of 4 times lower ($n \approx 1.2$ - 1.4). At a density of about 6 times lower than the standard solid density the experimental dependence changes its curvature.

The data obtained in [10] differs from those obtained in [7] and from this work because of a large systematic error in determining the volume and the resistive part of the voltage drop. In experiments [10], the wires were heated in glass capillaries such that the ratio of the inner diameter of the capillary to the initial wire diameter was 8. The sample volume was determined by recording the luminosity of the expanding plasma column through the thick capillary wall. At the early stage of the process when the wire vaporized the plasma column was nonuniform what is additional source of uncertainty in that measurement.

It can be seen from Fig. 5 that the theory developed in [12] yields resistivity that is close to that obtained in experiment 3 (and an analogous character of the dependence of the resistivity on the specific volume is predicted). The calculations carried out in [12] refer to the $T = 30$ kK isotherm. The temperature obtained in our calculations for the relative volume 20 in experiment 3 is about 36 kK. In this work the resistivity in the plasma region for experiment 3 is slightly lower (within 15%) compared to those in [6]. This is due to some changes in the EOS parameters. The reason was to mach better the critical point parameters obtained in [7].

It is of interest to estimate the characteristic density corresponding to the transi-

tion of tungsten from a metallic to dielectric state. According to the predictions made in [15], this transition is continuous and is not a first-order phase transition. The characteristic density is determined from the condition that the fraction of the classically accessible volume for valence electrons of the atoms of which the metal is formed amounts to about 30%. At a low temperature, the corresponding relative volume is approximately equal to 5. At a temperature of $T \approx I_1/3$ (where I_1 is the first ionization potential of tungsten), the metal-to-dielectric transition region should completely disappear. For tungsten, this temperature turns to be of about 30 kK. Hence, the knee observed in this work (experiment 3) seems to be unrelated to the theoretical predictions of [15], as far a temperature of about 30 – 34 kK is achieved near the knee and the corresponding relative volume reaches a value of 9.

6. CONCLUSION

The technique used here has allowed us realize homogeneous heating of a tungsten sample and measure its resistance during a continuous transition from condensed to gaseous state. The tungsten density decreased from the standard solid density to a density of 15 to 20 times lower. Using the technique, we have carried out experiments on a nearly isobaric heating of tungsten in a gaseous state. We have found that the conductivity-to-density ratio for tungsten in the liquid state remains almost constant at densities ranging from the melting point density down to that approximately 8 to 9 times lower than the normal solid density. At lower densities, the dependence of the conductivity on the density along the 30-kbar isobar changes its character: the conductivity becomes independent of density and approaches a nearly constant value. This change in the character of the dependence manifests itself as a knee in the corresponding experimental curve. As the pressure increases from 30 to 90 kbar, the density dependence of the conductivity near the knee becomes remarkably flatter and smoother. Nevertheless the main features observed at the low pressure persist at the maximal pressure applied.

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